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(54) Abstract Title
Spot cleaning compositions

(57) Spot cleaning compositions particularly useful for the localized cleaning of stains from garments and textiles comprise a surfactant, a solvent, an enzyme and xanthan gum as a thickening agent for the aqueous composition.

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IMPROVEMENTS IN OR RELATING TO ORGANIC COMPOSITIONS

The present invention is directed to improved spot cleaning compositions particularly useful for the localized cleaning of stains from garments and textiles.

The prior art has suggested many types of spot cleaning compositions which are intended to be used in the localized cleaning of stains on garments or textiles. Such spot cleaning is typically shortly performed prior to a subsequent laundering step wherein the garment or textiles are immersed in an aqueous bath.

The performance of these spot cleaning compositions is known to vary considerably, based on the types of stains which are to be removed, as well as on the type of textile from which a garment is made. Frequently, while known spot cleaning compositions are effective for many types of stains, they are not necessarily effective in the removal of a test soil, 'bandy black clay' which is notoriously difficult to remove.

A further shortcoming of many known art spot cleaning compositions is that they frequently need to be specifically formulated, or reformulated, for use in a specific class of container or dispensing device. Thus, a manufacturer of such a product may need to simultaneously produce several different formulations of one product, which is inconvenient and costly.

The present invention addresses and overcomes many of these shortcomings known in the prior art.

In a first aspect of the invention there is provided a thickened, aqueous cleaning composition which is particularly useful in the spot cleaning of stains and soils on garments and textiles which comprises:

a detersive surfactant selected from anionic, nonionic, cationic, amphoteric and zwitterionic surfactants as well as mixtures thereof, preferably at least one nonionic surfactant;

an organic solvent, preferably selected from alcohols, diols, glycols, glycol ethers, C_6-C_{18} methyl ester as well as N-methyl-2-pyrrolidone;

an enzyme constituent, preferably one which includes proteases;

a xanthan gum as the primary thickener constituent.

The compositions may optionally, but in some cases desirably include one or more further constituents including but not limited to: pH adjusting agents, pH buffering agents, chelating agents, hydrotropes, perfumes, perfume carriers, fluorescing agents, optical brighteners, colorants such as dyes, hydrotropes, enzyme stabilizers, builders, germicides, fungicides, preservative constituents, anti-oxidants, anti-corrosion agents, antistatic agents as well as other conventional additives known to the relevant art.

These compositions specifically do not include acrylate based thickeners such as those marketed under the tradenames CARBOPOL®, or ACCUSOL®, or clay based thickeners such as laponites.

The compositions of the invention desirably exhibit a relatively low viscosity under high shear conditions, and conversely, exhibit relatively high viscosity under low shear conditions. Such behavior ensures that the compositions may be used in both hand pumpable trigger spray dispensers, and in nozzled dispensers. In the former, high shear conditions exist at the pump, low shear conditions exist at the nozzle. The advantageous viscosity characteristics permit for the same formulation to be used in both of these types of dispensers, which are popularly used with this type of consumer product.

In a further aspect of the invention there is provided a process for the localized cleaning, viz., "spot cleaning" of a soiled or stained garment or textile which process contemplates the application to the soil or stained area of a cleaning effective amount of the compositions described herein. Desirably, such spot cleaning is carried out just prior to a subsequent laundering step.

The inventive compositions include one or more detersive surfactants selected from anionic, nonionic, cationic, amphoteric and zwitterionic surfactants and mixtures thereof. By way of non-limiting example, the useful anionic surfactants include soaps (also known as 'carboxylates') and the sulfated and sulfonated synthetic detergents. Exemplary soaps are generally the water soluble salts of saturated higher fatty acids of 8 to 18 carbon atoms each, and mixtures. The sulfated and sulfonated anionic surfactants include those having about 8 to about 26 and preferably about 10 to about 22 carbon

atoms to the molecule; and may optionally be alkoxylated. Examples of useful anionic surfactants include alkyl sulfates and their salts, such as sodium lauryl sulfate. Further exemplary detersive surfactants include amine oxides, particularly amine oxides in which the alkyl group has about 10-20, and preferably 12-16 carbon atoms, and can be straight or branched chain, saturated or unsaturated. The lower alkyl groups may include between 1 and 7 carbon atoms. Examples include lauryl dimethyl amine oxide, myristyl dimethyl amine oxide, and those in which the alkyl group is a mixture of different amine oxide, dimethyl cocoamine oxide, dimethyl (hydrogenated tallow) amine oxide, and myristyl/palmityl dimethyl amine oxide. Amphoteric detergents may also be used, including the salts of higher alkyl beta-amino propionic acids, e.g., sodium N-lauryl beta-alanine; the higher alkyl substituted betaines, such as lauryl dimethylammonium acetic acid; and the imidazoline type of amphoteric detergents. Exemplary cationic surfactants include both short chain alkyl, and long chain alkyl quaternary ammonium compounds. The former of these are typically associated with providing a germicicdal or sanitizing benefit to the compositions of which they form a part.

Preferably the inventive compositions include least one nonionic surfactant. Exemplary nonionic surfactants include the polyoxyethylene ethers of alkyl aromatic hydroxy compounds, e.g., the alkylated polyoxyethylene phenols, the polyoxyethylene ethers of long chain aliphatic alcohols, the polyoxyethylene ethers of hydrophobic propylene oxide polymers. Desirably the nonionic surfactants are one or more alcohol ethoxylates. Particularly useful are alcohol ethoxylates in the GENAPOL® "26-L" series which include C_{12}-C_{16} linear alcohols condensed with varying amounts of ethylene oxide. Particularly useful are those which are described in the Examples.

Desirably at least two different detersive surfactants are present, as mixtures of different surfactants are expected to provided a broader range of detergency. When the preferred linear alcohol ethoxylates are used, it is preferred that two (or more) be present, and that there be at least a 50% difference in the average moles of ethoxylation between these two. For example, where a first alcohol ethoxylate is used having an average of 4 moles of ethoxylation, then it is preferred that the second alcohol ethoxylate present have an average of at least 6 moles of ethoxylation.

The inventors have also found that the relative amounts of the preferred nonionic linear alcohol ethoxylates plays a contributing part to the ultimate viscosity of the formulation. Desirably, when two linear alcohol ethoxylates are used, and they exhibit at least a 50% difference in the average moles of ethoxylation, the respective weight ratios of the linear alcohol ethoxylate with the higher degree of ethoxylation to the linear alcohol ethoxylate with the lower degree should be within the respective ranges as exemplified by the Examples described below.

Desirably the detersive surfactant consists solely of one or more, especially two or more nonionic surfactants.

The detersive surfactant is present in amounts of from 0.001 – 25%wt. based on the total weight of the composition. Desirably the detersive surfactant is present in an amount of from 0.1 – 16%wt., more preferably from 1 – 12%wt.

The compositions of the invention also include at least one organic solvent, preferably at least one organic solvent selected from alcohols, diols, glycols, glycol ethers, C_6-C_{18} methyl ester as well as N-methyl-2-pyrrolidone. Useful organic solvents are those which are at least partially water-miscible such as alcohols (such as ethanol, n-propanol, isopropanol, the various isomers of butanol, etc.), diols, glycols (such as propylene glycol, hexylene glycol, etc.) water-miscible ethers (e.g. diethylene glycol diethylether, diethylene glycol dimethylether, propylene glycol dimethylether), water-miscible glycol ethers (e.g. propylene glycol monomethylether, propylene glycol mono ethylether, propylene glycol monopropylether, propylene glycol monobutylether, ethylene glycol monobutylether, dipropylene glycol monomethylether, diethyleneglycol monobutylether), lower esters of monoalkylethers of ethyleneglycol or propylene glycol (e.g. propylene glycol monomethyl ether acetate) all commercially available from Union Carbide, Dow Chemicals or Höchst. Also useful are C_6-C_{18} methyl esters (such as methyl caprylate-caprate, methyl laurate and methyl oleate; such as EMERY® and EMEREST® methyl esters, ex. Henkel) as well as N-methyl-2-pyrrolidone. Mixtures of organic solvents can also be used. Particularly preferred are the organic solvents illustrated in the Examples.

The organic solvent is present in amounts of from 0.001 – 20%wt. based on the total weight of the composition. Desirably the organic solvent is present in lesser amounts, desirably from 0.1 – 15%wt., more desirably from 1-15%wt.

The compositions include an enzyme constituent which includes one or more enzymes which are effective in the breakdown of certain stains and soils. Preferably the enzyme constituent includes proteases which are known to be effective in the breakdown of protein based stains, such as blood, mucus, grass, egg and gravy. Protein based stains are known to be particularly difficult to remove, but in the presence of an effective protease, they are hydrolized into peptides and may be more readily removed in a subsequent laundering step. Enzymes which are contemplated as useful include lipases, amylases, peroxidases, pectinases and the like. Combinations of two or more different enzymes may also be present, but it is preferred that at least proteases be present.

The enzyme constituent need be present in an effective amount. It will be recognized by skilled practitioners that the activity of the enzymes in a commercially available preparation may vary, and that such activity is very relevant in determining the amount of the commercially available preparation to be used in the inventive composition. Thus, one skilled in the art may determine by means of a few routine experiments what amount of a commercially available enzyme preparation is to be included in the inventive compositions. By way of non-limiting example, one commercially available preparation, SAVINASE® (ex. Novo Nordisk) is a protease based preparation which is advantageously present in amounts of from 0.001 – 5%wt. based on the total weight of the composition. Desirably, lesser amounts need be present, such as from 0.001 – 1%wt. Particularly preferred amounts are demonstrated in the Examples.

The inventive compositions include a xanthan gum as the primary thickener constituent. More specifically the inventive compositions do not include other thickeners based on acrylic polymers, or naturally occurring or synthetic clay thickeners. The inventors have surprisingly found that the inventive compositions provides two unique benefits. First, the use of xanthan gum as the thickener provides the beneficial viscosity characteristics which allows for the use of the same composition in both hand pumpable

trigger spray dispensers, as well as in nozzled dispensers. Second, the use of xanthan gum as the thickener also provides improved cleaning of stains, as opposed to similar compositions which substitute either an acrylic polymer or synthetic clay thickener in the place of the xanthan gum.

The xanthan gum typically needs to be present in only minor amount in order to provide a satisfactory thickening effect to the inventive composition. It is to be understood that the actual amount will vary on the ultimate viscosity characteristics which are desired, but advantageously amounts of from 0.001 – 10%wt. based on the total weight of the composition are considered to be generally effective. More desirably, the xanthan gum is present in lesser amounts, desirably from 0.01 – 8%wt. Of course higher amounts may be used where a thicker formulation is desired.

As is noted above, the compositions according to the invention are aqueous in nature. Water is added in order to provide to 100% by weight of the compositions of the invention. The water may be tap water, but is preferably distilled or soft water, and is most preferably soft water or deionized water. The inventive compositions desirably include at least 70%wt. water, more desirably include at least 80%wt. water.

Compositions of the invention may also include one or more optional constituents including, but not limited to: pH adjusting agents, pH buffering agents, chelating agents, hydrotropes, perfumes, perfume carriers, fluorescing agents, optical brighteners, colorants such as dyes, hydrotropes, enzyme stabilizers, builders, germicides, fungicides, preservative constituents, anti-oxidants, anti-corrosion agents, antistatic agents as well as other conventional additives known to the relevant art.

In certain particularly preferred embodiments, borax is included as an effective pH buffering constituent (ex. U.S. Borax Co.). This constituent is preferred for use as the buffer as it is believed to contribute to stablizing the enzyme constituent in the aqeuous compositions. Others however can be used as well, such as the citrates, citric acid, and the like.

In certain particularly preferred embodiments, a minor but an effective amount of a biocidal composition, which acts as a preservative, is also included. One such material

is DOWICIL® 75 (ex. Dow Chem. Co.) Others may be used as well, as long as they do not deleteriously effect the other constituents, especially the enzyme constituent.

Such constituents as described above as essential and/or optional constituents are per se, known to the art.

According to preferred embodiments, the compositions of the invention exhibit a viscosity in the range of from about 15,000 to about 62,000 centipoise when measured on a Brookfield viscometer, using a #2 spindle; 0.3 rpm, 75°F (23.8°C). Preferably the compositions exhibit a viscosity from about 20,000 to about 35,000 at these test conditions.

The compositions of the invention can be prepared in a conventional manner such as by simply mixing the constituents in order to form the ultimate aqueous cleaning composition. The order of addition is not critical, but it may be convenient to form a premixture of the organic solvents and the detersive surfactants at an elevated temperature, and to subsequently blend these into the remaining constituents which are dispersed in water. Conveniently, the xanthan gum is added as the last constituent.

The compositions of this invention may be packaged in any suitable container, such as an unpressurized bottle or in a pressurized container. They may be pressurized and made available in this form by means of the addition of a suitable propellant to the composition such as known hydrocarbon propellants including propane, butane, isobutane, and isopentane, halogenated hydrocarbon propellants including chlorodifluoromethane, difluoroethane dichlorodifluoromethane as well as pressurized gases such as carbon dioxide and nitrogen. Most desirably, the compositions are packaged and provided in a container especially a pressurized vessel or a manually operable pump which induces foaming of the composition as it is dispensed from the container.

Substrates which can be treated in accordance with this invention are textile fibers or filaments, and finished or fabricated fibrous articles such as textiles, and garments. The textiles include those made of natural fibers, such as cotton and wool, as well as those made of synthetic organic fibers, such as nylon, polyolefin, acetate, rayon, acrylic and polyester fibers.

The following examples illustrate the superior properties of the formulations of the invention and particular preferred embodiments of the inventive compositions. The terms "parts by weight" or "percentage weight" as well as "%wt." are used interchangeably in the specification and in the following Examples wherein the weight percentages of each of the individual constituents are indicated in weight percent based on the total weight of the composition, unless indicated otherwise.

Examples:

Illustrative exemplary formulations within the scope of the present inventive compositions are provided on Table 1 below, which are designated as "Example" or "Ex." formulations. Comparative examples are indicated as "C" formulations on Table 1.

Table 1						
	Ex.1	**Ex.2**	**Ex.3**	**Ex.4**	**Ex.5**	**Ex.6**
Genapol 26-L-60	4.5	5.0	5.5	9.0	9.0	3.1
Genapol 26-L-3	5.5	5.0	4.5	1.0	1.0	4.9
propylene glycol	7.0	7.0	7.0	7.0	7.0	8.0
Borax 10 mole	0.51	0.51	0.51	0.51	0.51	0.51
Dowicil 75	0.05	0.05	0.05	0.05	0.05	0.05
Kelzan ST	--	--	--	--	--	0.30
Keltrol T	0.30	0.30	0.30	0.40	0.70	--
Savinase 16.0L	0.41	0.41	0.41	0.41	0.41	0.41
Fragrance	0.1	0.1	0.1	0.1	0.1	0.1
di water	to 100	10 100	to 100	to 100	to 100	to 100

Table 1								
	C1	**C2**	**C3**	**C4**	**C5**	**C6**	**C7**	**C8**
Genapol 26-L-60	4.5	5.0	5.5	9.0	3.1	9.0	4.5	5.5
Genapol 26-L-3	5.5	5.0	4.5	6.0	4.9	1.0	5.5	4.5
propylene glycol	7.0	7.0	7.0	7.0	8.0	7.0	7.0	7.0
Borax 10 mole	0.51	0.51	0.51	0.51	0.51	0.51	0.51	0.51
Dowicil 75	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05
Laponite Clay	0.4	0.4	0.4	--	--	--	--	--
Carbopol 2623	--	--	--	0.58	0.15	--	--	--
Acusol 820	--	--	--	--	--	0.2	0.4	0.4
Savinase 16.0L	0.41	0.41	0.41	0.41	0.41	0.41	0.41	0.42
Fragrance	0.1	0.1	0.1	0.1	0.1	1.0	1.0	1.0
di water	to 100	10 100	to 100	to 100	to 100	to 100	to 100	to 100

It is to be understood that the amounts of the constituents are listed above are "as is" weights as supplied by the respective supplier. The identity of the individual constituents described in Table 1 above, their percentage by weight 'actives', as well as presently available commercial sources, are described in more detail in Table 2, below.

TABLE 2	
Genapol 26-L-60	alcohol ethoxylate (100%wt) (Clariant Corp.)
Genapol 26-L-3	alcohol ethoxylate (100%wt) (Clariant Corp.)
propylene glycol	propylene glycol (100%wt.) (Dow Chem.Co.)
Borax 10 mole	borax (U.S. Borax Co.)
Dowicil 75	preservative (Dow Chem. Co.)
Acusol 820	acrylate based thickener (30%wt. active) (Rohm & Haas Co.)
Laponite RD Clay	clay based thickener (Southern Clay Products)
Carbopol 2623	acrylate based thickener (30%wt.) (B.F. Goodrich)
Kelzan ST	xanthan gum based thickener (supplied at 100%, but dispersed in water to form a 1%wt. dispersion) (Kelco Co.)
Keltrol T	xanthan gum based thickener (supplied at 100%, but dispersed in water to form a 1%wt. dispersion) (Kelco Co.)
Fragrance	proprietary
Savinase 16.0L	protease containing, enzyme preparation (16 KNPU/gram according to Novo Nordisk test protocol for proteolytic activity) (Novo Nordisk Inc.)
di water	deionized water

The viscosity of certain of the compositions described on Table 1 were evaluated using a Brookfield viscometer, with a #2 spindle and a rotational speed of 0.3 rpm. The test was carried out at 75°F (23.8°C). All of the samples were evaluated 'as mixed'. The results of this test are illustrated on Table 3, below:

Table 3						
	Ex.1	Ex.2	Ex.3	Ex.4	Ex.5	Ex.6
viscosity, centipoise (#2 spindle; 0.3 rpm, 75°F)	55,000	35,000	30,000	20,000	60,000	28,000
viscosity, centipoise (#2 spindle; 3 rpm, 75°F)	7870	6280	5890	4160	EEEE	4430

'EEEE' indicated that the viscosity was higher than appropriate for the test conditions (spindle, rpm, temperature).

Table 3									
	C1	C2	C3	C4	C5	C6	C7	C8	SHOUT®
viscosity, centipoise (#2 spindle; 0.3 rpm, 75°F)	15,000	10,000	6600	1000	67,700	700	1700	5000	83,500
viscosity, centipoise (#2 spindle; 3 rpm, 75°F)	2300	1700	1690	–	–	140	1470	1420	9630

'–' indicates not tested,
the identity of SHOUT® is described below

Spot cleaning Testing:

Certain of the formulations described above were tested in order to evaluate their spot cleaning performance on stained textile swatches. This evaluation was in accordance with the protocol outlined in ASTM D-4265 (Reapproved 1998), using the standardized 'bandy black clay' as the test soil. This test soil and its source is defined in the protocol.

In the test, there were used test substrates of which unbleached cotton which did not have any surface treatment such as sizing, etc. These were obtained from Test Fabric Co. and are known standardized test swatches. For each formulation tested, there were six swatches used, each of which was treated as follows. To each of the swatches were applied five different stains, namely grape juice, bandy black clay, used motor oil, spaghetti sauce (RAGU®, Old World with Meat) and grass stains. The swatches were prepared in accordance with ASTM D-4265. The stains were allowed to dry. Subsequently, on each stain was used an aliquot of 2 ml. of a formulation to be evaluated, lightly rubbed three times from the reverse side of the swatch to distribute the forulation,

and within 1 minute were introduced into a washing machine. The washing bath contained 85 grams of a commercially available laundry detergent (TIDE®, ex. Procter & Gamble Co.) and washed for 12 minutes, at a wash bath temperature of 90 °F (32.2°C), followed by a cold water rinse, and spun to remove excess water. The laundered swatches were then removed, tumble dried in an domestic clothes dryer after which they were removed and SRI values evaluated as described below.

Due to the very poor viscosity characteristics of formulations containing laponite clays, these were not tested for stain removal performance.

For both of the tested formulations, solids removal, *viz.*, stain removal from the various soils was assessed quantitatively using a Hunter Lab colorimeter, which measured each of the following values: the lightness (" L_c ") of the unstained swatch; the lightness of the stained and subsequently washed swatch (" L_w "); redness-greenness of the unstained swatch(" a_c "), redness-greenness of the stained and subsequently washed swatch (" a_w "), yellowness-blueness of the unstained (" b_c ") swatch, and yellowness-blueness of the stained and subsequently washed swatch (" b_w "); each of these values measured as the amount of the standardized white light reflected from the fabrics. The quantitatively evaluated values were measured for the various tested fabric swatches and were used to calculate the Stain Removal Index (SRI) according to the equation:

$$SRI = 100 - [(L_c - L_w)^2 + (a_c - a_w)^2 + (b_c - b_w)^2]^{1/2}$$

and the results are reported on Table 4, below. Therein are reported the average values of the six stained swatches for each stain per formulation tested. The SRI value ranges from 0 to 100, with a value of 100 indicating complete soil removal.

Table 4

	grape juice	bandy black clay	used motor oil	spaghetti sauce	grass
Ex.1	88.34	81.43	85.50	93.12	94.17
Ex.3	88.68	92.42	85.28	92.63	94.39
Ex.5	89.16	92.33	83.48	94.06	93.85
C4	89.07	80.64	84.39	93.99	94.29
C7	88.34	81.43	85.50	93.12	94.17
Spray n' Wash®	88.39	84.19	85.66	93.34	92.43

SHOUT® 'push-pull' formulation	88.34	92.04	85.31	94.96	94.59

As can be seen from the results reported on Table 4 the reported SRI values indicate excellent stain removal performance of the inventive compositions, particularly with respect to removal of bandy black clay. Surprisingly, these results demonstrate that the inventive compositions demonstrate better removal of bandy black clay than similar compositions which substitute either an acrylic polymer or synthetic clay thickener in the place of the xanthan gum.

Table 4 also reports the stain cleaning performance of two presently commercially available spot-cleaning compositions, "Spray n'Wash®" (ex Dowbrands Inc.), and SHOUT® Laundry Stain Remover in a nozzled 'push-pull' bottle (ex S.C. Johnson & Son.) which indicates improved or comparable performance with these commercial products.

Claims:

1. A thickened, aqueous cleaning composition which is particularly useful in the spot cleaning of stains and soils on garments and textiles which comprises:
a detersive surfactant selected from anionic, nonionic, cationic, amphoteric and zwitterionic surfactants and mixtures thereof;
an organic solvent;
an enzyme constituent;
a xanthan gum as the primary thickener constituent;
and optionally one or more further constituents including but not limited to: pH adjusting agents, pH buffering agents, chelating agents, hydrotropes, perfumes, perfume carriers, fluorescing agents, optical brighteners, colorants such as dyes, hydrotropes, enzyme stabilizers, builders, germicides, fungicides, preservative constituents, anti-oxidants, anti-corrosion agents, antistatic agents

2. A thickened composition according to claim 1 wherein,
the detersive surfactant is selected from anionic and nonionic surfactants.

3. A thickened composition according to any preceding claim wherein,
the detersive surfactant includes at least one linear alcohol ethoxylate.

4. A thickened composition according to any preceding claim wherein,
the detersive surfactant includes at least two linear alcohol ethoxylates.

5. A thickened composition according to any preceding claim wherein,
the composition exhibits a viscosity of from about 15,000 to about 62,000 centipoise when measured on a Brookfield viscometer, using a #2 spindle; 0.3 rpm, 75°F (23.8°C).

6. A thickened composition according to any preceding claim wherein, the organic solvent is selected from alcohols, diols, glycols and glycol ethers.

7. A thickened composition according to any preceding claim wherein, the organic solvent consists solely of propylene glycol.

8. A thickened composition according to any preceding claim wherein, the enzyme constituent comprises a protease.

9. A thickened, aqueous cleaning composition which is particularly useful in the spot cleaning of stains and soils on garments and textiles substantially as described with reference to the Examples.

10. A process for the localized cleaning of a soiled or stained garment or textile which process comprises the step of:

applying to the soil or stained area of a cleaning effective amount of the compositions according to any preceding claim.

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Application No:	GB 9916492.3	**Examiner:**	Michael Conlon
Claims searched:	1 - 10	**Date of search:**	8 October 1999

Patents Act 1977
Search Report under Section 17

Databases searched:

UK Patent Office collections, including GB, EP, WO & US patent specifications, in:

UK Cl (Ed.Q): C5D (DHC, DHZ, D127)

Int Cl (Ed.6): C11D 3/22

Other: Online: WPI, EPODOC, PAJ

Documents considered to be relevant:

Category	Identity of document and relevant passage		Relevant to claims
A	US4532066	(Sterling)	1
A	US4855069	(Rhone Poulenc)	1

X	Document indicating lack of novelty or inventive step	A	Document indicating technological background and/or state of the art.
Y	Document indicating lack of inventive step if combined with one or more other documents of same category.	P	Document published on or after the declared priority date but before the filing date of this invention.
&	Member of the same patent family	E	Patent document published on or after, but with priority date earlier than, the filing date of this application.